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DEC 15 2005

IN THE UNITED STATES PATENT
AND TRADEMARK OFFICE

Applicant(s): Nozomu Takano et al
Serial No.: 10/771,260
Filed: February 4, 2004
For: PREPREG, METAL-CLAD LAMINATE, AND PRINTED CIRCUIT
BOARD OBTAINED FROM THESE
Art Unit: 1712
Examiner: Jeffrey B. Robertson

Honorable Commissioner of Patents
and Trademarks
Washington, D.C. 20231

DECLARATION UNDER 37 CFR 1.132

SIR:

I. I, Nozomu Takano, a first inventor of this case, declare
and say as follows.

I am one of the joint inventors of the present U.S.
Patent Application as identified above and understand the
English language. I studied the Official Action dated June 15,
2005 received in said application.

In order to clarify that the present invention is not
obvious over the invention of Babcock et al. (U.S. Patent No.
5,340,644) or Leibfried (U.S. Patent No. 5,451,637) in view of
Takano et al. (EP 0 837 090 A1), I have conducted comparative
experiments as mentioned below under my supervision.

II. Comparative experiments

An object of the experiments is to show the criticality

of a volume amount of the inorganic filler of the present invention to those of the references cited by the Examiner, which do not specifically mention the criticality thereof.

[Critical meaning of the filler in an amount of 25% by volume or more]

Experiment I

In the present invention, wire bonding properties are improved by increasing a surface hardness at high temperature region. To heighten the surface hardness, it is important that a modulus of elasticity of the substrate is high, and in the present invention, it could be accomplished "by formulating an inorganic filler in the resin composition". That is, by formulating an inorganic filler having less modulus of elasticity, modulus of elasticity of the whole resin composition can be effectively raised. Moreover, for improving the modulus of elasticity, it is necessary that the inorganic filler be so contained in the resins of the substrate in a "dense" state with a certain extent. In fact, as shown in Fig. 1 mentioned below, the modulus of elasticity is abruptly improved at a neighbor of 25% by volume. From this fact, by formulating the filler in an amount of 25% by volume or more, the surface hardness of the substrate is increased, and the wire bonding properties are also good.

Also, as shown in Fig. 2, by formulating the inorganic filler in an amount of 25% by volume or more, marked lowering in thermal expansion can be observed. In a composite material constituted by a resin(s)/an inorganic material(s) such as a substrate for a package, etc., due to the difference in thermal expansions, cracks are likely generated at the

interface thereof, and this is one of the problems for improving reliability. To improve this, it is important to lowering thermal expansion, and it can be found out that the means employed in the present invention was also effective for lowering thermal expansion.

Table 1 Relation between filling ratio and bending elasticity (filler: silica)

| Filling ratio (% by volume) | 0 | 20 | 30 | 40 | 45 |
|-----------------------------------|------|------|------|------|------|
| Modulus of elasticity (GPa) | 22.2 | 23.8 | 26.0 | 30.2 | 31.5 |

Table 2 Relation between filling ratio and thermal expansion (filler: calcined clay)

| Filling ratio (% by volume) | 0 | 25 | 40 | 50 |
|--------------------------------|------|------|------|------|
| Thermal expansion (ppm/°C) | 13.8 | 12.7 | 9.79 | 7.62 |

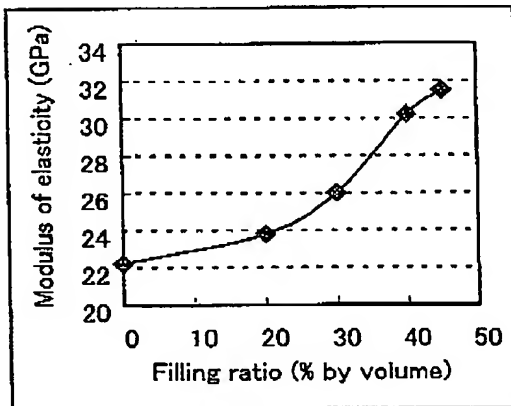


Fig. 1 Relation between filling ratio and bending elasticity (Filler: silica)

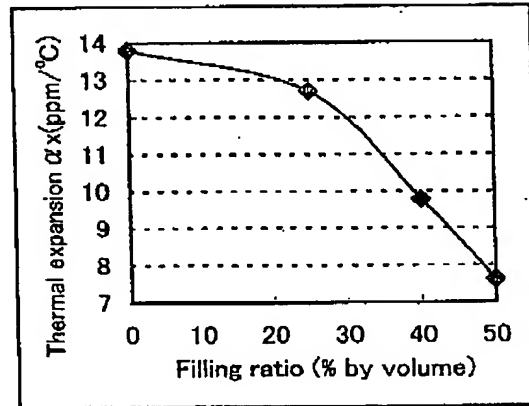


Fig. 2 Relation between filling ratio and thermal expansion (α x) (Filler: calcined clay)

(Experimental method)

1. Modulus of elasticity of a laminated board using silica as inorganic filler

To a glass flask equipped with a stirring device and a condenser was charged a solution comprising 20 g of dimethoxydimethylsilane, 25 g of tetramethoxy silane and 20 g of methanol, and to the solution were added 0.52 g of acetic acid and 17.8 g of distilled water, then, the resulting mixture was stirred at 50°C for 8 hours to synthesize a silicone polymer. An average of a siloxane recurring unit of the obtained silicone polymer was 25. To the silicone polymer solution was added methyl ethyl ketone to prepare a treating solution with a solid content of 10% by weight. To the treating solution was formulated 3200 g of silica as an inorganic filler, and the mixture was stirred at room temperature for 1 hour to prepare a solution containing the treated filler. The obtained solution containing the treated filler was heated to 50°C, and the inorganic filler in the solution was apportioned so that they became 0 parts by weight, 65 parts by weight, 112 parts by weight, 174 parts by weight and 214 parts by weight. To the respective solutions containing the respective treated filler were added resins and compounds in formulation amounts as shown below, and a solvent comprising methyl ethyl ketone and ethylene glycol monomethyl ether with a weight ratio of 50: 50 to prepare a resin composition varnish with a solid content of 70% by weight. Here, a ratio of the inorganic filler based on the total amount of the solid content of the resin composition was each 0% by volume, 20% by volume, 30% by volume, 40% by volume, and 45% by volume.

| | |
|---|--------------------|
| Brominated bisphenol A type epoxy resin | 50 parts by weight |
| ortho-Cresol novolac type resin | 50 parts by weight |
| Phenol-novolac resin | 42 parts by weight |
| 2-Ethyl-4-methylimidazole | 0.5 part by weight |

The respective varnishes prepared as mentioned above were impregnated into a glass cloth with a thickness of about 0.1 mm (Style 2116, E-glass), and heated and dried at 150°C for 5 minutes to obtain prepregs with a resin content of 48% by weight. Four sheets of this prepreg were laminated, and copper foils with a thickness of 18 µm were laminated on the both sides thereof, and a both surface copper clad laminated board were prepared under the press conditions of at 170°C, 90 minutes, and 4.0MPa.

The copper foils of the thus obtained both surface copper clad laminated board were removed from the whole surface by etching, and 3-point bending test was carried out. The 3-point bending test was carried out until the test piece has broken with a span of 20 mm, and a rate of a test speed of 0.8 mm/min, and a modulus of elasticity was calculated from an initial inclination of a load curve.

2. Thermal expansion of a laminated board using calcined clay as inorganic filler

In the same manner as in the above-mentioned 1, a solution containing a treated filler was prepared by using calcined clay in place of silica as an inorganic filler. The obtained solution containing the treated filler was heated to 50°C, and the inorganic filler in the solution was apportioned so that they became 0 part by weight, 103 parts by weight, 206 parts by weight and 309 parts by weight. To the respective

solutions containing the respective treated filler were added the same resins and compounds in the same formulation amounts as in the above 1, and a solvent comprising methyl ethyl ketone and ethylene glycol monomethyl ether with a weight ratio of 50: 50 to prepare a resin composition varnish with a solid content of 70% by weight. Here, a ratio of the inorganic filler based on the total amount of the solid content of the resin composition was each 0% by volume, 25% by volume, 40% by volume and 50% by volume.

The respective varnishes prepared as mentioned above were treated in the same manner as in the above 1 to prepare both surface copper clad laminated boards.

The copper foils of the thus obtained both surface copper clad laminated board were removed from the whole surface by etching, and a test piece was prepared so that a longitudinal direction of the glass cloth of the laminated board became 100 mm, and a lateral direction of the same became 10 mm. This test piece was heated from room temperature to 260°C with an elevation speed of 10°C/min using a continuous TMA (Thermomechanical analysis), and a thermal expansion between 60°C and 120°C was measured.

Experiment II

In the same manner as in Example 1 in the present specification except for changing an amount of the calcined clay as an inorganic filler from 1300 g to 800 g, a solution containing the filler was prepared. By using the solution containing the treated filler, prepreg and both surface copper clad laminated board were prepared in the same manner as in Example 1. A ratio of the inorganic filler based on the total solid content

of the resin composition was about 25% by volume.

With regard to the thus obtained both surface copper clad laminated board was evaluated surface smoothness, barcol hardness, wire bonding properties, solder heat-resistance and migration resistance were evaluated. The results are shown in the following with the results of Example 7 (ratio of filler: 40% by volume), Example 1 (ratio of filler: 37% by volume) and Example 9 (ratio of filler: 31% by volume) as well as Comparative example 5 (ratio of filler: 20% by volume).

| | Ratio of filler (%) | R_{max} | Barcol hardness 200°C | Wire bonding properties | | Solder heat-resistance | Migration resistance (hr) |
|-----------------------|---------------------|-----------|--------------------------|-------------------------|-----------------------|------------------------|---------------------------|
| | | μm | | Adhered ratio (%) | Adhesion strength (g) | | |
| Example 7 | 40 | 3 | 43 | 100 | 14 to 18 | OK | >500 |
| Example 1 | 37 | 5 | 38 | 100 | 12 to 16 | OK | >500 |
| Example 9 | 31 | 3 | 45 | 100 | 14 to 18 | OK | >500 |
| Additional Example | 25 | 5 | 36 | 100 | 12 to 16 | OK | >500 |
| Comparative example 5 | 20 | 15 | 20 | 73 | 7 to 10 | OK | >500 |

As can be seen from the above results, in the present invention, by making a ratio of the filler (% by volume) 25% by volume or more, surface roughness is as small as 5 μm or less, and the barcol hardness at 200°C was 36 or more. Also, an adhered ratio at the time of wire bonding at 200°C was 100% (failure ratio: 0%), and its adhesion strength is sufficiently high, whereby they have excellent wire bonding properties.

To the contrary, in the same of Comparative example, a ratio of the filler (% by volume) is 20% by volume (which is less than 25% by volume), the surface roughness is as large as

15 μ m, and the barcol hardness at 200°C was as low as 20. Also, an adhered ratio at the time of wire bonding at 200°C was 73% (failure ratio: 27%), and its adhesion strength is insufficient to as 7 to 10 g, whereby it is inferior in wire bonding properties to those of Examples.

III. Conclusion

I believe that the above results would indeed be surprising and could never be expected from the description of the cited references. Thus, I do not believe that the present invention is obvious over the references in combination cited by the Examiner.

IV. I further declare that all statements made herein of my own knowledge are true and that all statements made in information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001, of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Dated: _____

Nozomu Takano